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COHERENT SCATTERING OF LIGHT INTO HIGH FREQUENCY
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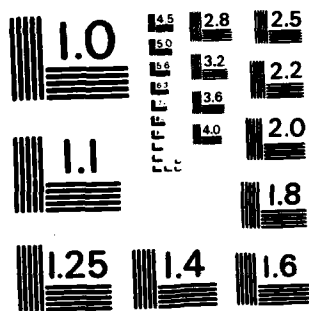
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COHERENT SCATTERING OF LIGHT INTO HIGH FREQUENCY RADIOWAVES

Annual
~~Final~~ Report

to

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March 31, 1983

J. Weber

University of Maryland, College Park, Maryland

and

University of California, Irvine, California



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Coherent Radiation Interaction, and Scattering by Atomic Nuclei in a Crystal

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ABSTRACT

The coherent radiation interaction, and scattering, by nuclei of a crystal for which each volume element has the same sign of the interaction with an incident beam, and for which the coupling of scatterers with each other is important, is computed.

Experiments are described which appear to verify the theory.

Introduction

The scattering of x-rays by solids has been well understood for many years. Each volume element of the solid has charges of both signs. Nonetheless the smaller mass of the electrons and the manner in which they are coupled to nuclei and to each other result in a scattering amplitude mainly contributed by the electrons.

The nuclei of a solid may also interact with radiation. Their coupling with each other is readily observed in nuclear magnetic resonance and other experiments. The lattice is a much more rigid structure than the electrons. Sufficiently great rigidity will be shown to lead to much stronger interaction with certain kinds of radiation.

First we will present a general discussion of the interaction of two four current densities. This will be applied to a single scatterer with a number of possible sites.* Exchange of momentum by a single scatterer with several possible sites will be shown to have features which suggest a strong interaction mechanism for N tightly bound particles on N sites.

*Certain stationary states require an atom to be on more than one site. For example the nitrogen atom in ammonia occupies two potential minima, on each side of the plane of the hydrogen atoms, with equal probability.

INTERACTION OF FOUR CURRENT DENSITIES

Let us consider the S matrix for interaction of two four current densities¹ given by

$$S = \frac{1}{\hbar c} \int \langle F | \bar{\psi}_s \Gamma \psi_s \bar{\psi}_I K \psi_I | 0 \rangle d^4x \quad (1)$$

$\langle F |$ is the final state, $|0\rangle$ is the original state. $\bar{\psi}_s$ is a creation operator for scatterer S, $\bar{\psi}_I$ is a creation operator for incident particle I. ψ_s and ψ_I are the corresponding annihilation operators. Γ and K are position independent operators.

The operators $\bar{\psi}_s$ and $\bar{\psi}_I$ are represented by the following expansions²

$$\begin{aligned} \bar{\psi}_s &= \sum_n \sum_j \psi_{sin}^* (\bar{r} - \bar{r}_n) a_{jn}^\dagger \\ \bar{\psi}_I &= \frac{1}{\sqrt{V}} \sum_k \bar{U}_{Ik} e^{-\frac{i}{\hbar} \bar{p}_{Ik} \cdot \bar{r}} d_k^\dagger \end{aligned} \quad (2)$$

In (2) \bar{r} is the position three vector, a_{jn}^\dagger is a creation operator for the state with wavefunction ψ_{sin}^* , n refers to the n^{th} scattering site. d_k^\dagger is a creation operator for an incident particle with known momentum \bar{p}_{Ik} . U_{Ik} is an incident particle spinor.

SINGLE SCATTERER ON N SITES

Suppose further that there are N sites in a solid material. For the states ψ_{sin} , harmonic oscillator states are selected. Consider the case of a single scatterer.

The single scatterer can be prepared in a harmonic oscillator ground state with equal probability to be on any one of the N sites. The original scatterer state for the single scatterer is taken to be

$$\frac{1}{\sqrt{N}} \sum_{n=1}^{n=N} a_{on}^{\dagger} \left| \begin{array}{c} \text{VACUUM} \\ \text{STATE} \end{array} \right\rangle \quad (3)$$

Let us assume that the scatterer position probability distribution $\psi_{sin}^* \psi_{sin}$ is not changed by the scattering. Therefore the effect of the scattering can only be a phase shift such that each final scatterer state $(\psi_{sin})_F$ is related to the original state $(\psi_{sin})_O$ by

$$(\psi_{sin})_F = (\psi_{sin})_O e^{i\Delta p_r x^r / \hbar} \quad (4)$$

(4) implies that each component in the momentum decomposition of the scatterer is shifted by the three momentum $\Delta \vec{p}$, corresponding to momentum exchange $\Delta \vec{p}$.

For a harmonic oscillator wavefunction centered at radius vector \vec{r}_n ,

$$\psi_{sin} = \left(\frac{\alpha}{\pi} \right)^{3/2} e^{-\alpha^2 |\vec{r} - \vec{r}_n|^2} \quad (5)$$

In (5) α specifies the volume occupied by the particle.

We assume spin zero scatterers, $\Gamma = 1, (1), (2), (3)$, and (4) give

$$S = \frac{\bar{U}_{IF} K U_{IO}}{\hbar c V N} \int \left(\frac{\alpha}{\pi} \right)^{3/2} \sum_{n=1}^{n=N} e^{-\alpha |\bar{r} - \bar{r}_n|^2 + \frac{i}{\hbar} (\bar{p}_{IO} - \bar{p}_{IF} - \Delta \bar{p}) \cdot \bar{r}_n} d^3x \quad (6)$$

In (6) \bar{p}_{IO} and \bar{p}_{IF} are the original and final 4 momenta of the incident particle, respectively. As $\alpha \rightarrow \infty$ (6) becomes

$$S \rightarrow \frac{\bar{U}_{IF} K U_{IO}}{\hbar c V N} \sum_{n=1}^{n=N} e^{\frac{i}{\hbar} (\bar{p}_{IO} - \bar{p}_{IF} - \Delta \bar{p}) \cdot \bar{r}_n} \int e^{\frac{i}{\hbar} (\bar{E}_{IO} - \bar{E}_{IF} - \Delta E) t} dt \quad (6A)$$

(6) indicates the possibility of exchanging the entire momentum $\Delta \bar{p}$ at any of the possible N sites at which the single scatterer may be found.

For momentum conservation, the sum in (6) approaches N. The possibilities for exchange of energy and momentum do not appear to severely restruct the solid angle into which an incident particle would be scattered.

COHERENT MOMENTUM TRANSFER PHASE SHIFT INTERACTIONS

Suppose now that we have N tightly bound scatterers. If an incident particle interacts with one scatterer, its strong coupling with all other nuclei might be expected to affect the interaction with an incident particle in a profound way. Consider (6). As already noted, the entire momentum $\Delta \bar{p}$ can be exchanged at any site without possibility of identifying the site of the scattering. With sufficiently strongly coupled particles momentum transfer at a single site is immediately exchanged with all other particles, with no possibility of identifying the site at which the scattering occurred.

For N tightly bound scatterers the original state is selected as

$$a_{o1}^{\dagger} a_{o2}^{\dagger} a_{o3}^{\dagger} \cdots a_{oN}^{\dagger} \left| \begin{array}{c} \text{VACUUM} \\ \text{STATE} \end{array} \right\rangle \quad (7)$$

For nuclei in a solid, the wavefunctions of different scatterers will not overlap to a significant degree, and the symmetry of the wavefunction need not be considered.

For exchange of momentum Δp_{μ} at the j^{th} site, $\bar{\psi}_s$ in (2) must be replaced by

$$\bar{\psi}'_s = \psi_{soj}^* a_{oj}^{\dagger} e^{-\frac{i\Delta p_{\mu} x^{\mu}}{\hbar}} + \sum_{i \neq j} \psi_{s oi}^* a_{oi}^{\dagger} \quad (8)$$

We may write (8) in a more illuminating form by adding $\psi_{soj}^* a_{oj}^{\dagger}$ to the last term and subtracting it from the first term to give

$$\bar{\psi}'_s = \psi_{soj}^* a_{oj}^{\dagger} \left[e^{-\frac{i\Delta p_{\mu} x^{\mu}}{\hbar}} - 1 \right] + \sum_{\text{ALL } N} \psi_{s oi}^* a_{oi}^{\dagger} \quad (8A)$$

In (8A) the last term gives the probability amplitude for the possible process where no momentum is exchanged at any site. The first term then gives the contribution to the amplitude for exchange Δp_{μ} at the j^{th} site. Since we are assuming strong coupling of nuclei to each other with no possibility of identifying the scattering site, we must sum only the first term in (8A) over all possible sites.

This gives, for coherent momentum transfer phase shift scattering

$$S = \frac{\bar{U}_{IF} K U_{IO}}{\hbar c V} \int \sum_{n=1}^{n=N} \left(\frac{\alpha}{\pi}\right)^{3/2} e^{-\alpha |\bar{n} - \bar{n}_n|^2 + \frac{i}{\hbar} (p_{IO} - p_{IF} - \Delta p)_r x^r} d^4x \quad (9)$$

SCATTERING CROSS SECTIONS

Suppose now that we have nuclei in a cubic crystal with N identical cells each with length a. For these assumptions the S matrix (9) for initial and final states in which the harmonic oscillator quantum numbers are the same, is given by

$$S = \bar{U}_{IF} K U_{IO} X Y Z T \left(\frac{1}{\hbar V}\right) \quad (10)$$

with

$$X = \sum_{n=1}^{n=N^{1/3}} e^{\frac{i}{\hbar} (p_{IO} - p_{IF} - \Delta p)_x X_n - \frac{1}{\alpha} \left(\frac{p_{IO} - p_{IF} - \Delta p}{2\hbar}\right)^2_x}$$

In (10), $X_n = n a$, with corresponding definitions for Y and Z. As before, a is a parameter specifying the width of the harmonic oscillator wavefunction.

$$T = \frac{\sin \left[\frac{(E_{IF} - E_{IO} + E_{SF} - E_{SO}) \tau}{2\hbar} \right]}{\left[\frac{E_{IF} - E_{IO} + E_{SF} - E_{SO}}{2\hbar} \right]} \quad (11)$$

E_{IF} and E_{SF} are the final state energies of the incident particle and ensemble of scatterers respectively, E_{IO} and E_{SO} are the corresponding original energies.

The scattering cross section is given by σ , with

$$\sigma = \sum \frac{V(S-1)^2}{c\tau} = \frac{V}{(2\pi)^6 c\tau\hbar^8} \int \left| \bar{U}_{IF} K U_{IO} XYZT \right|^2 dp_s dp_I \quad (12)$$

In (12) $d\bar{p}_S$ is the element of momentum space for the final state of the ensemble of scatterers, $d\bar{p}_I$ is the element of momentum space for the final state of the incident particle. T in (11) and (12) is a function of the momentum variables in X , Y , and Z . The integration (12) is carried out in the following way:

The length L of the crystal is given by $L = aN^{1/3}$, to a very good approximation we may evaluate

$$\frac{L}{2\pi\hbar} \int X^2 dp_{sx} = \frac{L}{2\pi\hbar} \int \left[\frac{\sin \left(\frac{N^{1/3}a [p_{Io} - p_{If} - \Delta p]_x}{2\hbar} \right)}{\sin \left(\frac{a [p_{Io} - p_{If} - \Delta p]_x}{2\hbar} \right)} \right]^2 e^{-\frac{2}{a} \left(\frac{p_{Io} - p_{If} - \Delta p}{2\hbar} \right)^2} dp_{sx} = N^{2/3} \quad (13)$$

Combining (13), (12) and (11) then gives

$$\sigma = \frac{N^2 (\bar{U}_{IF} K U_{IO})^2}{(2\pi)^3 c\hbar^5 \tau} \int T^2 d\bar{p}_I = \frac{N^2 (\bar{U}_{IF} K U_{IO})^2}{(2\pi)^3 c\hbar^5 \tau} \left(T_{p_I} \right)^2 \frac{d|p_I|}{dE} dE d\Omega_I \quad (14)$$

with $E = E_I + E_S$, $d\Omega_I$ is the element of solid angle into which the incident particle is scattered.

In the center of mass system

$$\frac{d|p_I|}{dE} = \frac{E_{IF} E_{SF}}{c^2 p_I (E_{IF} + E_{SF})} \quad (15)$$

(14) is integrated over E first

$$\sigma = \frac{N^2 (\bar{U}_{IF} K U_{I0})^2}{4\pi^2 c^3 \hbar^4} \int \bar{P}_I \frac{E_{IF} E_{SF}}{(E_{IF} + E_{SF})} d\Omega_I \quad (16)$$

(13) implies that

$$d\Omega_I \sim \left[\Delta p + \frac{2\hbar\pi}{N^{1/3}a} \right] \frac{1}{(\bar{P}_{I0})^2}$$

The integral (16) is over all values of Ω which approximately conserve energy and momentum as implied by the integrations (13) and (14). It can be shown for the case of zero rest mass particles, that a large volume of phase space meets these criteria. (16) may approach N^2 times the cross section of a single particle on one site.

The result (16) was obtained for the very simple cubic model. A similar result may be obtained for any very tightly bound group of scatterers even if these are not arranged in a perfect periodic lattice. For the more general case we may define

$$f(\bar{p}_{IF} - \bar{p}_{I0} - \Delta\bar{p}) = \int e^{\frac{i}{\hbar}(\bar{p}_{IF} - \bar{p}_{I0} - \Delta\bar{p}) \cdot \bar{r}'} \psi_{so}^*(\bar{r}') \psi_{so}(\bar{r}') d\bar{r}' \quad (17)$$

$$R = f(\bar{p}_{IF} - \bar{p}_{I0} - \Delta\bar{p}) \sum_{n=1}^{n=N} e^{\frac{i}{\hbar}(\bar{p}_{IF} - \bar{p}_{I0} - \Delta\bar{p}) \cdot \bar{r}_n} \quad (18)$$

In terms of (17) and (18) the S matrix is

$$S = \frac{\bar{U}_{IF} K U_{I0} R T}{\hbar V} \quad (19)$$

R may be evaluated in the following way. In (18) consider the sum

$$\sum_{n=1}^{n=N} e^{\frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot \bar{r}_n} \quad (20)$$

and express it as the product of factors involving X_n, Y_n, Z_n

$$\sum_{n=1}^{n=N_x} e^{\frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot X_n} \sum_{n=1}^{n=N_y} e^{\frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot Y_n} \sum_{n=1}^{n=N_z} e^{\frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot Z_n} \quad (21)$$

the object

$$X' = \sum_{n=1}^{n=N_x} e^{\frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot X_n} \quad (22)$$

is the sum of N_x unit vectors. The last one in the sum makes an angle

$$\theta_{N_x} = \frac{i}{\hbar} (\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \cdot X_{N_x} \quad (23)$$

with the first. The increments in angle are not equal, however the sum is given approximately by

$$X' = N_x \frac{\sin \frac{\theta_{N_x}}{2}}{\frac{\theta_{N_x}}{2}} \quad (24)$$

Similar expressions result for Y' and Z' , and

$$R = X' Y' Z' f(\bar{p}_{If} - \bar{p}_{Io} - \Delta \bar{p}) \quad (25)$$

The phase space integrals then give a result similar to (16).

DISCUSSION

The large cross sections (16) result from three very important assumptions. The ensemble of scatterers is assumed to be infinitely stiff, and recoils in the same manner as a single elementary particle on the N sites. Expression (8) then states that a final ensemble state differs from an initial state only in the phase factor $e^{\frac{i \Delta p_{\mu} x_{\mu}}{\hbar}}$. This phase factor is crucial for obtaining a large cross section because it may enormously increase the solid angle into which scattering occurs.

Suppose first that the phase factor $e^{\frac{i \Delta p_{\mu} x_{\mu}}{\hbar}}$ is absent -- as in the published solutions for potential scattering -- in which energy but not momentum is conserved.³ The absence of Δp_{μ} may enormously decrease the value of (16), because under this condition (13) implies (center of mass system),

$$\frac{N^{1/3} a (p_{I0} - p_{If})}{2\hbar} \ll \pi \quad (26)$$

(26) then limits the solid angle into which scattering may occur, expression (16), to

$$\Delta \Omega < \left(\frac{2\hbar\pi}{N^{1/3}a} \right) \frac{1}{p_{I0}^2} = \left[\frac{2\pi(\text{DE BROGLIE WAVELENGTH OF INCIDENT PARTICLE})}{\text{LENGTH OF SCATTERER ARRAY}} \right]^2 \quad (27)$$

The limitation of the Ω integration by (27) results in an extremely small cross section. This limit disappears when the phase factor $e^{i \frac{\Delta p \cdot x}{\hbar}}$ is included, for Δp the same value for all scatterers.

This follows from the modification of (27) as a result of the collective momentum transfer phase shift, to

$$\Delta \Omega < \Delta p + \frac{2\hbar\pi}{N^{1/3}a} (p_{I0})^2 \quad (28)$$

For large N , if $\Delta p \rightarrow p_{I0}$, (28) is enormously greater than (27). The cross section (16) and transition probabilities are correspondingly increased.

The second assumption is that the ensemble consists of highly localized particles which do not, therefore, have well defined momenta. It can be shown that if the momenta of all scatterers are precisely known before and after the interaction with the incident beam, the total cross section (and transition probability) will be very small.

The third assumption is that the sign of the interaction is the same in all volume elements. For electromagnetic radiation incident on a solid this requires an applied nearly uniform field to obtain essentially the same polarization in all volume elements. For the neutrino field, the universal Fermi interaction has the same sign for all particles of the solid.

INTERACTION OF PHOTONS WITH AN ENSEMBLE OF NUCLEAR MOMENTS

Several experiments are being carried out to verify this theory. One involves light which is incident on a nearly perfect crystal which is very nearly transparent. Interaction is with the nuclear moments. The nuclei are tightly coupled to each other. An applied constant magnetic field H_0 gives a net magnetic polarization. The magnetic moment μ is given by

$$\mu = g \mu_0 \hbar I \quad (29)$$

In (29) g is the gyromagnetic ratio, I is the spin vector in units of \hbar , μ_0 is the nuclear magneton given in terms of the nuclear mass M , electron charge e , and speed of light c , by

$$\mu_0 = \frac{e}{2 M \hbar c} \quad (30)$$

The Maxwell vector potential operator is given, in Coulomb gauge by

$$A_i = \sqrt{\frac{8\pi\hbar c}{V}} \sum \frac{1}{\sqrt{2k}} \sum_{m=1}^{m=3} \left(a_m(k) \epsilon_i^m e^{ik \cdot x} + a_m^\dagger(k) \epsilon_i^m e^{-ik \cdot x} \right) \quad (31)$$

In (32), $a_m^\dagger(k)$ and $a_m(k)$ are creation and annihilation operators, respectively, for photons. ϵ_i^m are a pair of orthonormal unit vectors, in a plane perpendicular to \vec{k}

For interaction of electromagnetic radiation with nuclear moments, the S matrix is given by

$$S = \frac{g \mu_0}{c} \int \langle F | \psi_s^\dagger \psi_s \epsilon^{abc} I_a \frac{\partial A_b}{\partial x^c} | 0 \rangle d^4x \quad (32)$$

In (32) ϵ^{abc} is the three space Levi Civita tensor density. It is zero if two indices are equal and unity if all indices are different.

$\epsilon^{123} = +1$ and changes sign on the interchange of any pair of indices.

For N scatterers in harmonic oscillator ground states, (32) is evaluated as

$$S = g \mu_0 \sqrt{\frac{4\pi \hbar c}{V}} \int \sum_{n=1}^{n=N} \left(\frac{\alpha}{\pi}\right)^{3/2} e^{-\alpha |\bar{n} - \bar{n}_n|^2 + \frac{i}{\hbar} (p_{x0} - \Delta p)_\mu x^\mu} \langle F | \eta \cdot I | 0 \rangle d^4x \quad (33)$$

In (33) \bar{n} is an appropriate unit vector defined by (32), in the direction of the incident light magnetic field. (33) may be written in terms of the integrals X , Y , Z , T defined earlier with (10), as

$$S = g \mu_0 \sqrt{\frac{4\pi \hbar c}{V}} X Y Z T \langle F | \eta \cdot I | 0 \rangle \quad (34)$$

Following the procedures of (12), (13) and (14), the cross section for absorption, or emission, is computed to be

$$\begin{aligned} \sigma &= 4\pi \mu_0^2 k q^2 \hbar N_r^2 \tau^{-1} |\langle F | \eta \cdot I | 0 \rangle|^2 \int T^2 \rho(E) dE \\ &= 8\pi^2 k \mu_0^2 q^2 \hbar^2 \rho(E) |\langle F | \eta \cdot I | 0 \rangle|^2 N_r^2 \end{aligned} \quad (35)$$

N_r is the difference between the number of spins parallel and antiparallel to the polarization field.

(35) may be written in terms of the fine structure constant α_f , the Compton wavelength λ_{CN} of the scatterers, the angular frequency ω_L of the incident light, and the nuclear magnetic resonance angular frequency in the applied time independent magnetic field ω_{NMR} as

$$\sigma = 2\pi^2 \alpha_f \left(\frac{\omega_L}{\omega_{NMR}} \right) \lambda_{CN}^2 \left| \langle F | \vec{r} \cdot \vec{I} | 0 \rangle \right|^2 N_p^2 \quad (35A)$$

Expression (35A) will usually exceed the projected area of the crystal, implying total extinction in passage of light through the first portion of the crystal traversed by the light. The dynamic diamagnetism has not been considered. This will reduce the light intensity at the nuclei, and limit the total absorption. It is possible that more precise measurements and an improved formulation of the theory will lead to a theory and measurements of the dynamic diamagnetism.

COHERENT ABSORPTION OR STIMULATED EMISSION ISSUES

The theory given earlier in equations (10)-(14) considered an incident particle with momentum \vec{P}_{I0} scattered into momentum \vec{P}_{IF} with momentum exchange $\Delta\vec{P}$. For the experiments reported here the incident particle may be absorbed and \vec{P}_{IF} is zero, or there may be stimulated emission. Here again the single scatterer exchange $\Delta\vec{P}$ is required to give a large cross section. Without the exchange $\Delta\vec{P}$, the sum of the exponential terms (10) will in general be very small.

Conservation of Momentum and Energy

For the process being considered, a photon is absorbed or created by the ensemble of scatterer magnetic moments. The entire ensemble recoils and its change of momentum Δp is balanced by the gain or loss of the photon momentum. Since the mass of the magnetic moments is very large, the recoil energy will be very much smaller than the photon energy,

$$E_{\text{RECOIL}} \ll \hbar \omega \quad (36)$$

Energy may be conserved if the spin system state is changed. The interaction will not be coherent over all N particles if the interaction changes the spin states of certain ones because these could then be identified as causing the interaction.

Here it is assumed that all interacting particles are in the same kind of quantum state and all are changed in the same way by the interaction. Suppose the magnetic moments are associated with spin $\frac{1}{2}$ particles. Every particle is assumed to have the spin state $\psi_0 = \begin{bmatrix} a_s \\ a_b \end{bmatrix}$ which is changed by the interaction to $\psi_F = \begin{bmatrix} a'_s \\ a'_b \end{bmatrix}$. To conserve energy it is necessary that

$$N [|a_s|^2 - |a'_s|^2 - |a_b|^2 + |a'_b|^2] \mu H_0 = \hbar \omega \quad (37)$$

A study of the integrations (33), (34), and (35) indicates that energy exchange as implied by (37) may occur in more than one way, without significant reduction of the total cross section. Each site interacts with the incident light, and a process in which momentum is exchanged at one site, and energy exchanged at many other sites simultaneously, leads to the large value (35). It can also be imagined that both energy and momentum are exchanged at a single site, and other interactions distribute the energy to the ensemble of nuclear spins.

Net Exchange of Energy

In spectroscopy and quantum electronics the interaction of radiation with a large number of particles in thermal equilibrium is usually described by the density matrix with random phases. For a particle with two quantum states U_1 and U_2 , with energies E_1 and E_2 , an equivalent description is to assume that N_1 are in the state U_1 , N_2 are in the state U_2 , and

$$N_2 = N_1 e^{-(E_2 - E_1)/kT} \quad (38)$$

If spontaneous emission is not important and the transition probability for stimulated emission is W_{12} , the net power exchanged is P_{net} , given by

$$P_{\text{net}} = W_{12} (N_1 - N_2) \hbar \omega_{12} \quad (39)$$

For the coherent process being considered here all particles are assumed to be in the same spin state. For spin $\frac{1}{2}$ the wavefunction for each particle is

$$\psi = a_1 \psi_1 + a_2 \psi_2 \quad (40)$$

The instantaneous net power exchanged is P_{net}^I

$$P_{\text{net}}^I = N \frac{d}{dt} \left[|a_1|^2 E_1 + |a_2|^2 E_2 \right] \quad (41)$$

Unitarity requires

$$|a_1|^2 + |a_2|^2 = 1 \quad (42)$$

(41) and (42) give

$$P_{NET}^I = N(E_1 - E_2) \frac{d}{dt} |a_y|^2 \quad (43)$$

(42) and (43) then give

$$P_{NET}^I = \frac{N}{2} (E_1 - E_2) \frac{d}{dt} (|a_y|^2 - |a_z|^2) \quad (44)$$

If the system remains in thermal equilibrium

$$|a_y|^2 = |a_z|^2 e^{-\frac{E_{L1}}{kT}} \quad (45)$$

the average net power is then

$$\langle P_{NET} \rangle = \frac{N}{2} (E_1 - E_2) \frac{d}{dt} \left[|a_y|^2 \left(1 - e^{-\frac{E_{L1}}{kT}} \right) \right] \quad (46)$$

Density of States and Matrix Elements

For the experiment in progress, a crystal of Lithium Fluoride interacts with light from a Helium Neon Laser. The transmitted light is measured with a photometer outside of the magnetic field. Changes are observed as the magnetic field is varied. (Figure 1) We may expect the density of states $\rho(E)$ in (35) to be given by

$$\rho(E) = \frac{1}{\hbar \omega_{NMR}} \quad (47)$$

In order to complete the calculation for the expected coherent absorption of light by an ensemble of magnetic moments we must evaluate the squared matrix element

$$|\langle F | \mu \cdot I | 0 \rangle|^2 \quad (48)$$

(48) is the square of the matrix element of the component of spin parallel to the magnetic field of the incident light.

EFFECTS OF AN APPLIED RADIOFREQUENCY FIELD AT THE NUCLEAR MAGNETIC RESONANCE FREQUENCY

Consider an isolated particle with spin $\frac{1}{2}$ having the wavefunction (40). Application of a radiofrequency magnetic field normal to the constant magnetic field at the exact resonance frequency will modify (40). Integration of the Schrodinger equation for interaction of a magnetic moment with the fields

gives

$$\psi = \begin{vmatrix} e^{-\frac{i\omega_{NMR}t}{2}} \left[a_{1s} \cos \frac{\omega_1 t}{2} - i a_{2s} \sin \frac{\omega_1 t}{2} \right] \\ e^{\frac{i\omega_{NMR}t}{2}} \left[-i a_{1s} \sin \frac{\omega_1 t}{2} + a_{2s} \cos \frac{\omega_1 t}{2} \right] \end{vmatrix} \quad (40A)$$

In (40A) $\omega_1 = \frac{\omega_{NMR} H_{\text{RADIOFREQUENCY}}}{H_0}$

The expectation values of the X, Y, and Z components of the nuclear spin are calculated from (40A) to be

$$\langle C_z \rangle = \langle |a_{1s}|^2 - |a_{2s}|^2 \rangle \cos \omega_1 t + i \langle a_{2s}^* a_{1s} - a_{1s}^* a_{2s} \rangle \sin \omega_1 t \quad (49)$$

$$\langle C_x \rangle = \langle (|a_{1s}|^2 - |a_{2s}|^2) \sin \omega_{NMR} t \sin \omega_1 t + i \langle a_{1s}^* a_{2s} - a_{1s} a_{2s}^* \rangle \sin \omega_{NMR} t \cos \omega_1 t \rangle \quad (50)$$

$$+ \langle a_{1s}^* a_{2s} + a_{1s} a_{2s}^* \rangle \cos \omega_{NMR} t$$

$$\langle C_y \rangle = \langle |a_{2s}|^2 - |a_{1s}|^2 \rangle \cos \omega_{NMR} t \sin \omega_1 t + i \langle a_{1s} a_{2s}^* - a_{1s}^* a_{2s} \rangle \cos \omega_{NMR} t \cos \omega_1 t \rangle \quad (51)$$

$$+ \langle a_{1s}^* a_{2s} + a_{1s} a_{2s}^* \rangle \sin \omega_{NMR} t$$

ω_{NMR} is very large, ω_1 is very small. Since the incident light magnetic field is parallel to the time independent magnetic field, $\langle C_z \rangle$ is believed to dominate the optics experiments discussed here in accordance with (48). $\langle C_x \rangle$ and $\langle C_y \rangle$ are usually observed in nuclear magnetic resonance.

Experiments

Red light from a Helium Neon Laser was employed with the apparatus of Figure 1. The interaction of this light with a lithium fluoride crystal was studied, as a function of applied fields, at 4.2 Kelvin. A silicon "solar" cell A served as photometer. The "conetic" shield reduced the magnetic field at the photometer to values sufficiently small to guarantee that changes in magnetic field did not significantly affect the photometer calibration.

The laser light was polarized with magnetic field of the light parallel to the magnetic field of a Varian electromagnet. Filters were employed to attenuate the 3.3 micron infra red light emitted by the laser. A lens served to diffuse the light so that the beam cross section was comparable with the area of the crystal. A neutral density filter reduced the intensity of the light so that approximately 10^{-12} watts illuminated the crystal.

A magnetic field of approximately 8,000 Gauss was applied and immediately this reduced the intensity of the transmitted light by approximately a factor of 2. Then on a time scale of hours, the intensity changed, undergoing oscillations as shown in Figure 2. Eventually, in some cases after more than 20 hours, the intensity dropped to a very low value, to less than ten per cent of the incident light before the magnetic field was applied. There are changes in the dimensions as the liquid level of helium changes. Allowing for this, it appears very likely that the "equilibrium" intensity - shown in Figure 2, after 20 hours, is less than thirty

per cent of the zero magnetic field intensity.

Refilling was required at least once in 24 hours. The enormous vibration levels which accompany transfer of liquid helium result in phonon densities sufficiently great to again excite the kind of oscillations shown in Figure 2.

Removal of the magnetic field again resulted in a long period relaxation, exceeding 6 hours, shown in Figure 3, back to the small attenuation levels.

Similar results were obtained with a sapphire crystal as shown in Figure 4.

With no applied radiofrequency fields, no evidence for a resonance in absorption of light was found as a function of the applied magnetic field.

The process described by (32) is an absorption of photons. The observed decrease in intensity might also be understood in terms of a higher order elastic scattering process. To check this possibility, a second photometer B was employed as shown in Figure 1. The crystal was cut and polished along a plane parallel to its axis and the polished face was covered by a silicon "solar" cell. The second photometer then observes light scattered at right angles to the incident rays. If the observed decrease in the direct light to photometer A is the single photon coherent absorption of equation (35), the outputs A and B should both decrease. If the observed decrease of

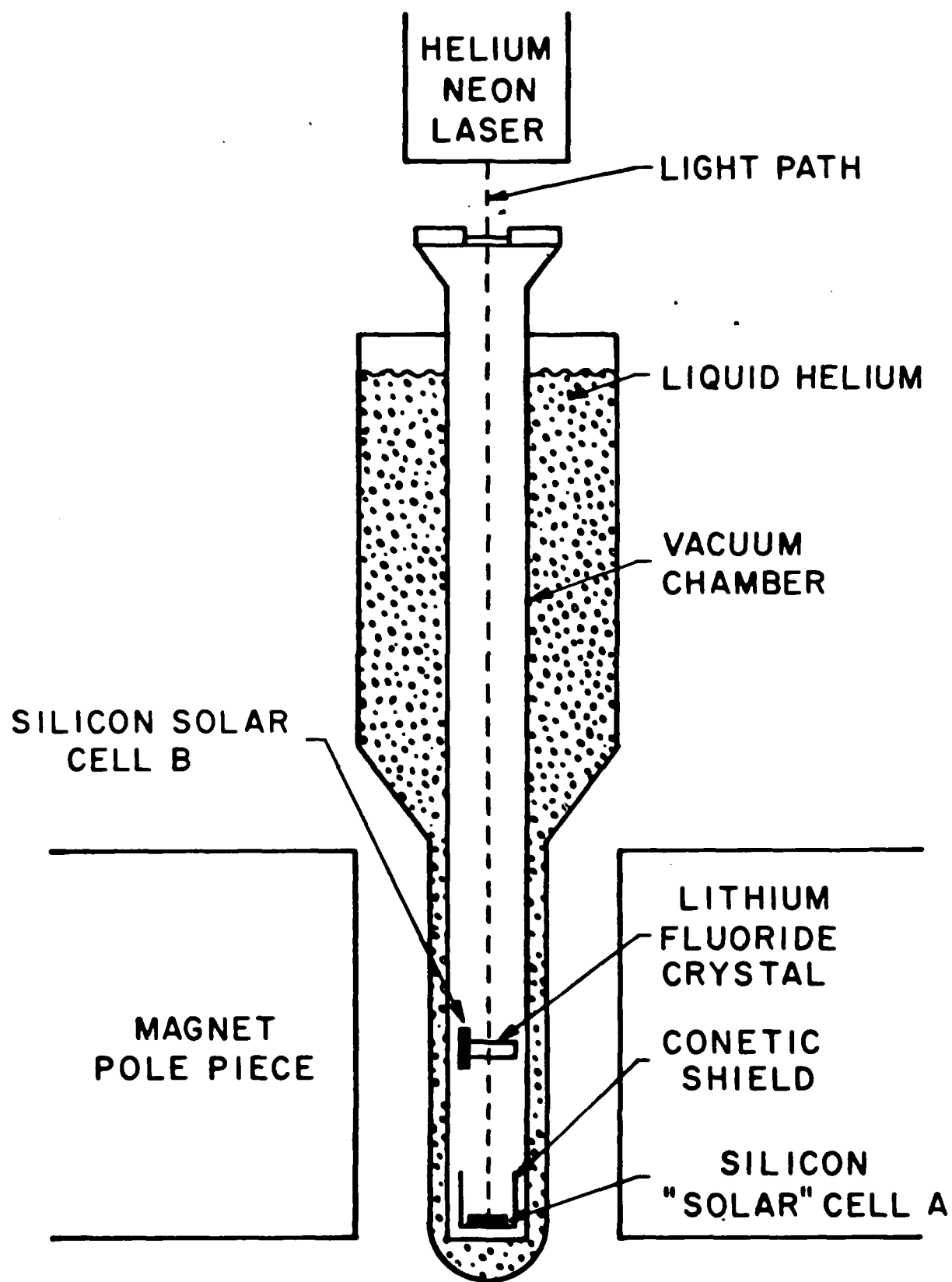
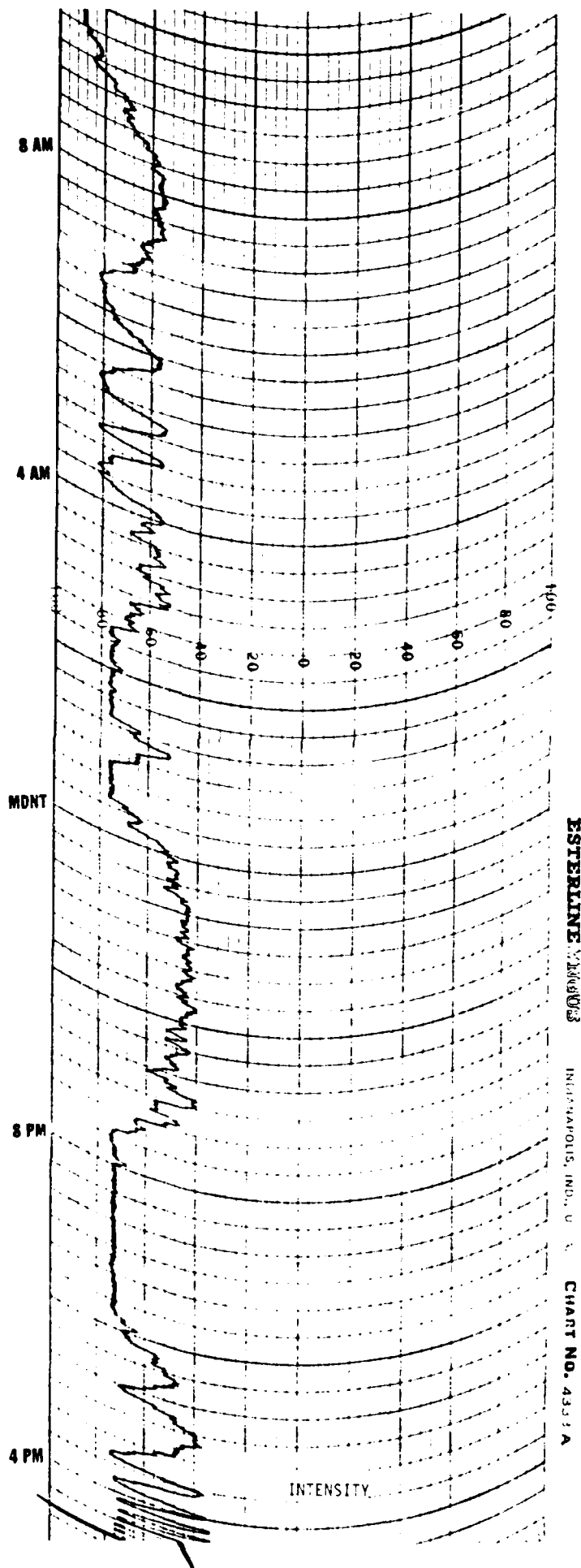


FIGURE 1

TIME IN HOURS

Figure 2



TIME IN HOURS

Figure 3

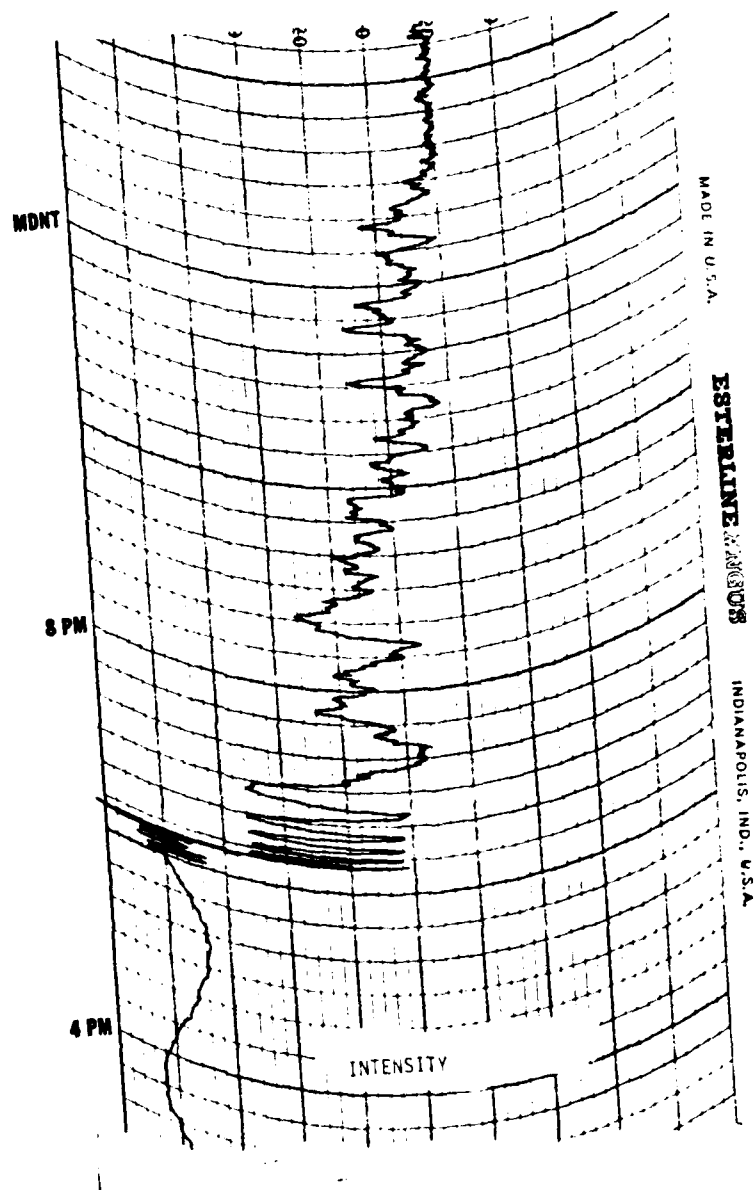
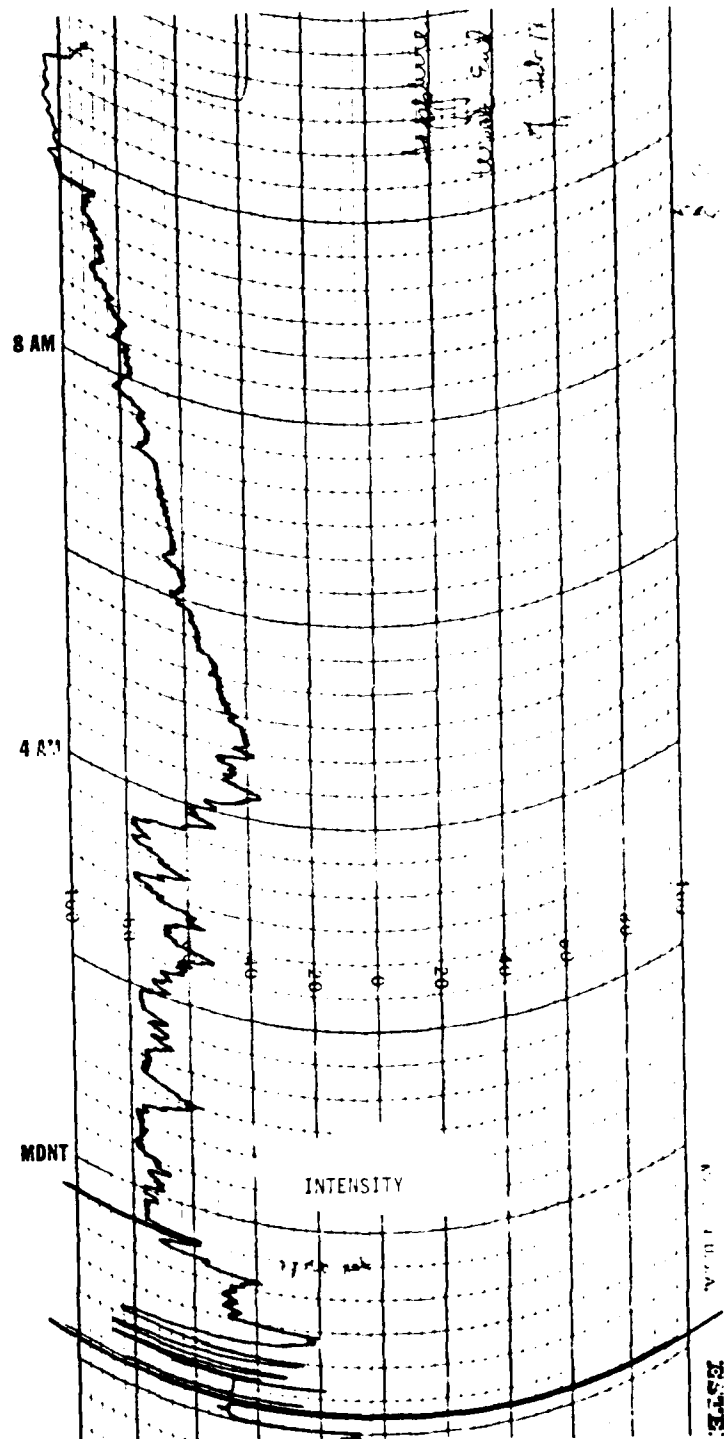


Figure 4



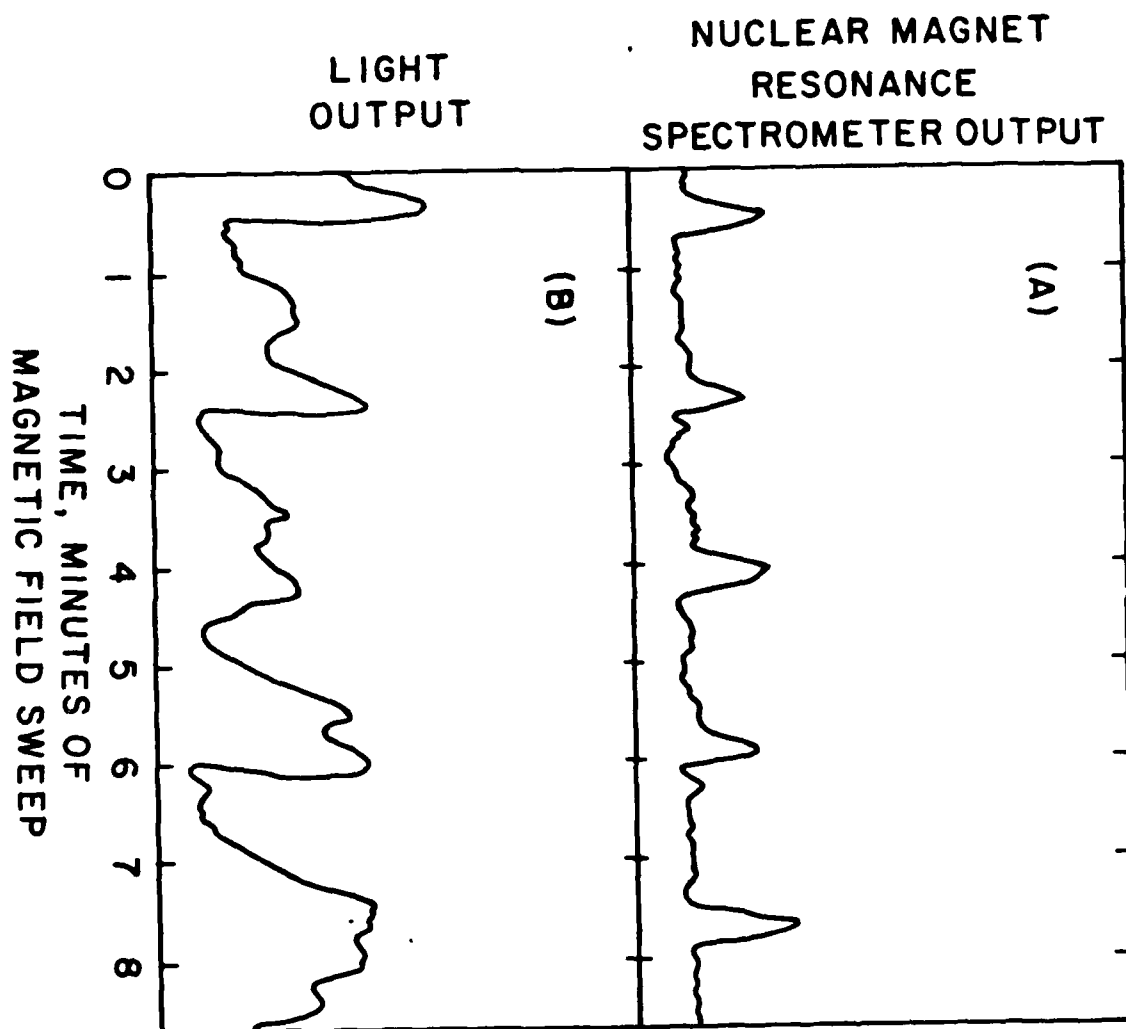


Figure 5

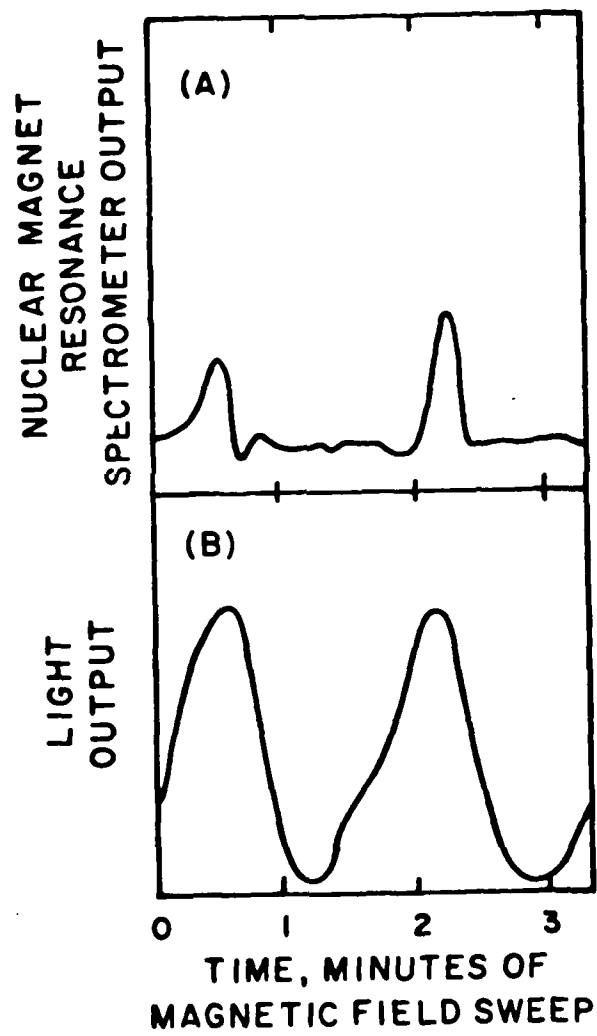
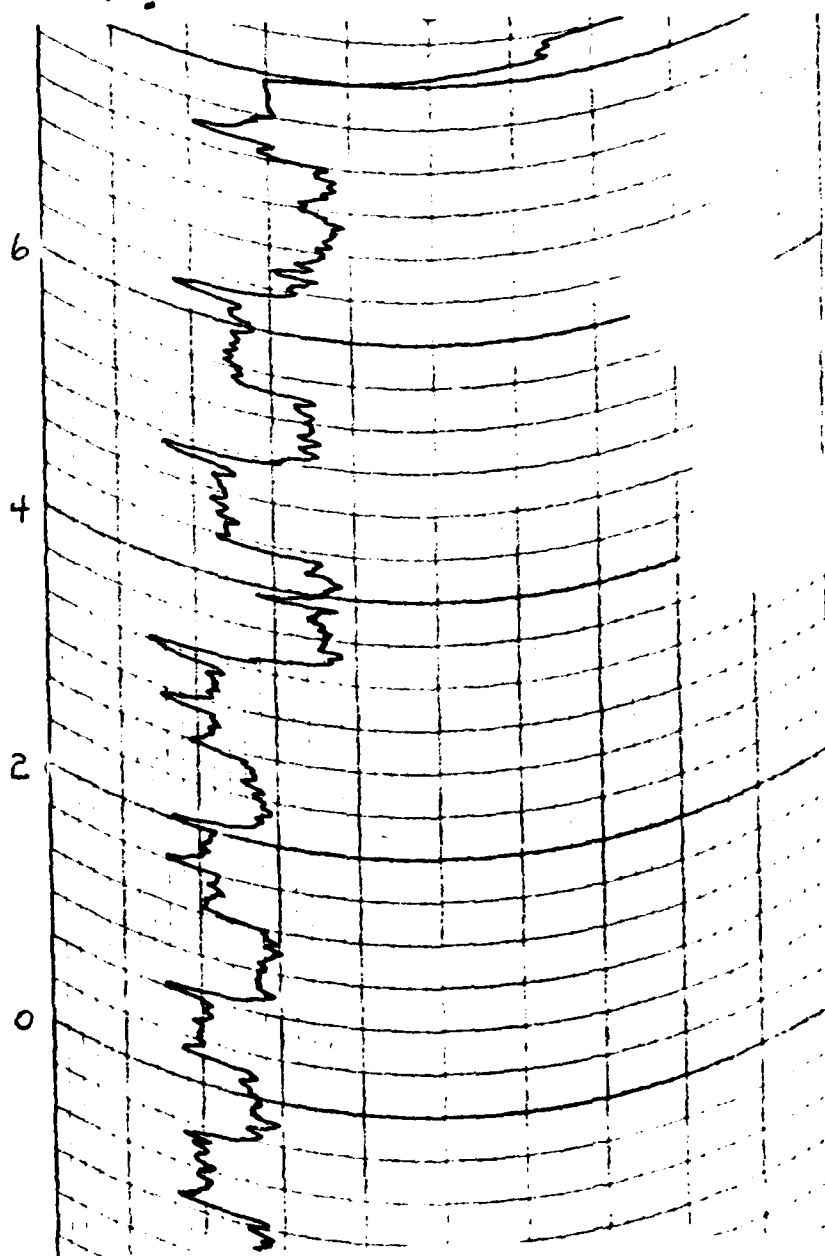


Figure 6

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TIME IN MINUTES

Figure 7

NMR RESONANCE INTENSITY

intensity in A is associated with elastic scattering, a decrease in A should be accompanied by an increase in B. All observations gave decreases in B when the direct light to A was observed to decrease. With reduction of the applied magnetic field, both photometer outputs increased. Within limits of experimental error the two photometer outputs always changed in the same direction by approximately the same fractional amounts.

Possible Heating Effects

A spin 1/2 system has specific heat C_v given by

$$C_v = \frac{1}{2T} \frac{NE}{\left(e^{\frac{E}{kT}} + 1\right)} \approx \frac{Nk}{4} \left(\frac{E}{kT}\right)^2 \approx 10^{-7} \quad \text{Joules per degree}$$

at 4K for the Fluorine nuclei in the crystal for an 8,000 Gauss field. Since the intensity of the light is five orders smaller, no thermal effects are expected. To explore this aspect the light source was turned off for an hour, after the system appeared to be in equilibrium. Restoring the light intensity to the earlier value reproduced the earlier result.

NUCLEAR MAGNETIC RESONANCE INTERACTIONS

Two lithium fluoride crystals were employed, both furnished by Harshaw. Both had been irradiated by x rays to permit easier grinding and polishing. One had been annealed. It was colorless. The spin lattice relaxation time for the Fluorine atoms was measured and found to be approximately three hours. The crystal which had not been annealed, appeared to be very light yellow, and measurements gave a spin lattice relaxation time approximately 40 minutes. Allowing for the difference in relaxation times, both crystals responded to light and radiofrequency fields in the same way.

To further explore the issue of correlations, the radiofrequency field was modulated at one hertz. The light output was amplified by a synchronous detector switched at the one hertz modulation frequency. Figure 5 shows the correlations. The upper trace is the recorded nuclear magnetic resonance output as the applied magnetic field was slowly swept through resonance. The lower trace is the light output from the synchronous detector at the same time. Figure 6 is for the same kind of data with signal averaging over six complete cycles to improve the signal to noise ratio. A radiofrequency field considerably less than required for saturation was employed. The observed correlations for this relatively small radiofrequency field are roughly one percent of the light output.

At other times the magnitude of the correlations varied considerably, depending on the spin state history.

It was then decided to search for correlations in the output of the nuclear magnetic resonance spectrometer with the incident light. Such correlations were found to be large at certain values of incident light intensity. At a power $\sim 10^{-9}$ watts from the helium neon laser, opening and closing the shutter produced the large changes in radiofrequency output shown in Figure 7, for the spectrometer tuned exactly to the Fluorine resonance. The spin state history

was again an important factor in the magnitudes of the correlations. At certain times there were nuclear spin slow heating effects (as interpreted from the NMR line heights) when the light was turned on. A thermistor mounted on the crystal indicated that the surface temperature of the crystal was unaffected by the light.

DISCUSSION OF THE EXPERIMENTS

Figures 2, 3, and 4 suggest that the absorption of light is modulated with approximately constant amplitude and monotonically increasing period until an equilibrium or quasistationary state is reached. One way (but not the only way) to understand these phenomena is to consider expressions (49) and (35). Since the magnetic field of the incident light is parallel to the time independent magnetic field, (35) suggests that the absorption cross section is proportional to $|\langle \epsilon_z \rangle|^2$.

(49) implies that harmonic excitation of the spin system with monotonically decreasing amplitude will indeed cause the period ω_1 to decrease, in accordance with the observations. Such excitation may be initiated by sudden changes in applied magnetic field or large phonon density, causing large fluctuations in spin energy. These may provide the excitation mechanism, as the system relaxes to equilibrium.

The wide range of the response of the light transmission to nuclear magnetic resonance radiofrequency radiation, and the equally wide range of response of the nuclear magnetic resonance line intensity to light might be understood from the following considerations. Soon after cooldown the spin system is relaxing to 4 Kelvin equilibrium. The spin lattice coupling is relatively strong and this facilitates energy exchange needed for heating of the spin system by an applied radiofrequency field, with large resultant change in polarization density.

After several days at 4 K the spin system is very cold and the spin lattice coupling is relatively weak. Under these conditions the spins act as though they are isolated, with response to a radiofrequency field given by (40A). (49) implies that the correlations $\langle a_i^\dagger, a_{2s} \rangle$, and $\langle a_i, a_{2s}^\dagger \rangle$ may contribute large changes to the total cross section.

ACKNOWLEDGEMENT

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CONCLUSION

Experiments suggest that large fractions of 6328 Å light incident on a crystal may be absorbed as a result of interaction with nuclear moments in consequence of momentum transfer.

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- ² Quantum Mechanics Non Relativistic Theory, Landau and Lifshitz, Volume 3, page 215
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FIGURE CAPTIONS

- Figure 1 Interaction of Laser Light with a Crystal
- Figure 2 Intensity of Transmitted Light Versus Time After Cooldown and Application of 8000 Gauss Magnetic Field for Lithium Fluoride Crystal
- Figure 3 Intensity of Transmitted Light Versus Time After Removal of 8000 Gauss Magnetic Field
- Figure 4 Intensity of Transmitted Light Versus Time After Cooldown and Application of 8000 Gauss Magnetic Field for Sapphire Crystal
- Figure 5 Correlation of Transmitted Light with Nuclear Magnetic Resonance Sweep
- Figure 6 Correlation of Transmitted Light with Nuclear Magnetic Resonance Sweep, Averaging over Six Complete Cycles.
- Figure 7 Correlations of Nuclear Magnetic Resonance Absorption with Incident Light

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